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STIMULATED RAMAN AND CONCENTRATION SCATTERING

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Because of a change-over from semi-annual to quarterly technical progress reports, effective June 1970, this technical report actually covers the progress since April 1, 1970. Since the summer of 1969, the main technical objective of this program has been the understanding of the process of stimulated concentration scattering. This is a scattering of intense light beams in a fluid mixture caused by concentration fluctuations at constant pressure and temperature. Since the atmosphere is a mixture of different gases, such processes might, in principle, affect the propagation of intense light beams in the atmosphere. Fortunately, the theory predicts that the concentration scattering in the atmosphere is negligible compared to Rayleigh scattering caused by temperature fluctuations and reorientation of anisotropic molecules. The theory predicts that stimulated concentration scattering could only become of comparable importance in a gaseous mixture where the constituents have a very large difference in mass and in polarizability. Appendix 1 contains a detailed analysis which was completed during this reporting period. This material has also been accepted for publication in the Physical Review.

The long term experimental effort of the past two years has also come to fruition during this period. The experimental demonstration of stimulated concentration scattering has been achieved in a mixture of SF_6 and helium gas. This subject matter is described in more detail in Appendix 2. It has been submitted for publication in Physical Review Letters and was presented in a paper read at the International Conference on Quantum Electronics in Kyoto, Japan, September 1970.

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APPENDIX A

THEORY OF STIMULATED CONCENTRATION SCATTERING[*]

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ABSTRACT

From the general formulation of the coupling of a laser wave and a scattered light wave with density, temperature and concentration fluctuations in a binary fluid mixture, an expression for the gain g_c of stimulated concentration scattering is derived. It is shown that this gain is usually much smaller than the gain for stimulated Brillouin scattering, g_B. However, in gaseous mixtures at relatively low pressures, with a large difference in polarizability as well as in mass between the two components, g_c can become larger than g_B. A large difference in polarizabilities increases the coupling of light to the concentration fluctuations, while a large difference in mass produces a pronounced increase in damping of a sound wave. The calculated values for g_c/g_B are compared with experimental results.

I. Introduction

Spontaneous light scattering from fluids has a long history. The spectral triplet in the scattered light spectrum, consisting of a central (Rayleigh) peak and a Brillouin doublet, is well known.[1,2] The low lying excitations of the medium involved in these scattering processes are the thermal diffusion and the acoustical phonon modes respectively. In a binary mixture of fluids the Rayleigh cross section for the central component can be very much enhanced, because the contribution from concentration fluctuations can be larger than that from temperature fluctuations. There are, of course, many other types of excitations which contribute to the scattered light spectrum. Examples are rotational motions or librations of anisotropic molecules (inelastic Rayleigh wing scattering) and vibrational excitations or optical phonon modes (Raman scattering). All of these spontaneous scattering processes have been studied with renewed vigor and much greater precision by means of gas laser beams during the past decade.[2,3]

In principle there are stimulated scattering processes associated with each spontaneous process.[4] The intensity in a scattered mode can be amplified exponentially with a gain coefficient proportional to the incident laser intensity. If this gain coefficient is larger than the unavoidable (linear) absorption and spontaneous scattering losses, some preferred scattered light modes may build up to very high intensities. Usually the stimulated process with the lowest threshold is dominant and may deplete the laser intensity before other processes can be stimulated. Due to differences in a transient response it is sometimes possible to excite two or more stimulated processes with a high intensity laser pulse.

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Since the response of low-lying hydrodynamic excitations is slow, they are best stimulated in relatively long laser pulses. The results of a steady state gain theory are valid in the limit that the spectral width of the laser is smaller than the spontaneous line width divided by the gain coefficient.

Stimulated Brillouin and Raman scattering are most frequently observed, and their relative steady state and transient characteristics have been studied in detail. Stimulated Rayleigh wing scattering in liquids from anisotropic molecules is also well established.[4]

Stimulated scattering associated with the sharp central Rayleigh peak is more difficult to demonstrate experimentally without ambiguity. It is characterized by a small frequency shift and is usually difficult to stimulate by the short pulses from Q-switched lasers. The thermal Rayleigh scattering from temperature fluctuations induced by absorption has been demonstrated convincingly,[5,6] and the thermal scattering induced by the electro-caloric effect in a non-absorbing fluid has been discussed in several papers by Fabelinskii and coworkers.[6]

Two experiments have been reported which claim to have demonstrated stimulated concentration scattering. One of these was concerned with a binary mixture of liquids,[7] the other with a mixture of gases.[8] It is the purpose of this paper to present a systematic investigation, analyzing the relative importance of the stimulated scattering from density, temperature and concentration fluctuations. In section II we present the relevant dynamical equations of a fluid mixture and discuss their range of validity. In section III the coupling mechanisms with the electromagnetic fields are analyzed. The concentration scattering can easily be made

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to dominate the thermal scattering, but the ever present Brillouin scattering usually has a much higher gain. The steady state solutions for stimulated concentration and Brillouin scattering from a binary fluid mixture are given in section IV. It is shown in section V that the stimulated concentration scattering can become dominant only in selected situations, e.g. in a binary gas mixture at relatively low pressure with two components with large differences in mass and polarizability. Numerical calculations on the competition between stimulated Brillouin and concentration scattering are compared with recent experimental results.[7-9] The agreement is satisfactory, but our calculations show some doubt on the correctness of the interpretation that stimulated concentration scattering was responsible for the earlier observations.[7,8]

II. Review of the Dynamical Equations in a Fluid Mixture

Consider a mixture of two fluids.[10] Let there be N_1 molecules of mass m_1 and N_2 molecules of mass m_2 per unit volume. The chemical potentials of the two species are μ_1 and μ_2 respectively. The differential free energy may then be expressed as

$$d F = - S d T - P d V + \mu_1 dN_1 + \mu_2 dN_2 \quad . \tag{1}$$

The mass density is

$$\rho = N_1 m_1 + N_2 m_2 = N[c'm_1 + (1-c')m_2] \quad , \tag{2}$$

where N is the total number of particles per unit volume and c' is the relative number concentration of the first species. It is related to the relative mass concentration c by,

$$c = c' \ [c' + (1-c') \ (m_2/m_1)]^{-1} \tag{3}$$

and

$$\frac{\partial c}{\partial c'} = \left(\frac{\partial c'}{\partial c}\right)^{-1} = \frac{m_1 m_2}{[m_1 c' + m_2(1-c')]^2} = \frac{[m_2 c + (1-c)m_1]^2}{m_1 \ m_2} \quad . \tag{4}$$

If pressure, temperature and mass concentration are taken as independent variables, the total differential of the Gibbs free energy per unit mass may be written as,

$$d \Phi = - s d T + v d P + \mu d c \quad , \tag{5}$$

where the chemical potential of the mixture per unit mass μ has been introduced,

$$\mu = (\mu_1/m_1) - (\mu_2/m_2) \quad . \tag{6}$$

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The diffusion current may be expressed as,

$$\underline{i} = \alpha \nabla \mu - \beta \nabla T \tag{7}$$

where the last term represents the thermal diffusion. The heat current is given by,

$$\underline{q} = \left(\mu + \frac{\beta T}{\alpha}\right) \underline{i} - \kappa \nabla T . \tag{8}$$

The last term represents the heat current in the absence of diffusion. These convection equations are valid if the relative variation of the thermodynamic quantities over a mean free path of the molecules is small.

The chemical potential μ is expanded as a function of P,T and c and the coefficients of diffusion, thermal diffusion and barodiffusion are introduced by the following relations,

$$D = \frac{\alpha}{\rho}\left(\frac{\partial\mu}{\partial c}\right)_{T,P} \quad ; \quad X = \frac{\kappa}{\rho\, c_P}$$

$$\frac{\rho K_T D}{T} = \alpha\left(\frac{\partial\mu}{\partial T}\right)_{c,P} + \beta \quad ; \quad \kappa = \gamma - \frac{\beta^2 T}{\alpha} \tag{9}$$

$$K_P = P\left(\frac{\partial v}{\partial c}\right)_{P,T} \bigg/ \left(\frac{\partial\mu}{\partial c}\right)_{P,T} = - \frac{P\left(\frac{\partial\rho}{\partial c}\right)_{P,T}}{\rho^2\left(\frac{\partial\mu}{\partial c}\right)_{P,T}} \quad .$$

For a dilute gas, obeying the ideal gas law, the chemical potentials of the individual species have the form,

$$\mu_1 = k_B T \ln c' + \psi_1(T) \tag{10}$$

$$\mu_2 = k_B T \ln(1-c') + \psi_2(T) ,$$

where the ψ's are functions of temperature only.

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With these relations one finds,

$$\left(\frac{\partial \mu}{\partial c}\right)_{T,P} = \frac{k_B T}{c(1-c) [m_2 c + (1-c)m_1]} \tag{11}$$

and

$$K_P \left(\frac{\partial c'}{\partial c}\right) = \frac{N (m_2 - m_1) \; c'(1-c')}{\rho} \qquad . \tag{12}$$

The fundamental hydrodynamic equations for the two component fluid mixture, which express the conservation of mass, momentum, energy and number of particles of individual species may be written in the form,

1) Continuity equation (conservation of mass)

$$\frac{\partial \rho}{\partial t} + \text{div}(\rho \, \underset{\sim}{V}) = o \tag{13}$$

2) Navier - Stokes equation (conservation of momentum)

$$\frac{\partial \underset{\sim}{V}}{\partial t} + (\underset{\sim}{V} \cdot \text{grad}) \; \underset{\sim}{V} = -\frac{1}{\rho} \; \text{grad} \; P + \frac{\eta}{\rho} \; \nabla^2 \underset{\sim}{V} + \frac{1}{\rho} \; (\tfrac{1}{3} \eta + \zeta) \; \text{grad div} \; \underset{\sim}{V} \tag{14}$$

3) Thermal diffusion equation (conservation of energy)

$$\frac{\partial T}{\partial t} - \frac{K_T}{c_P} \left(\frac{\partial \mu}{\partial c}\right)_{P,T} \; \frac{\partial c}{\partial t} + \frac{T}{c_P} \left(\frac{\partial s}{\partial P}\right)_{T,C} \; \frac{\partial P}{\partial t} = \chi \; \nabla^2 T \tag{15}$$

4) Concentration diffusion equation (conservation of particle number of individual species)

$$\frac{\partial c}{\partial t} = D[\nabla^2 c + \frac{K_T}{T} \; \nabla^2 T + \frac{K_P}{P} \; \nabla^2 P] \tag{16}$$

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In this macroscopic thermodynamic approach the transport coefficients are regarded as constants, to be obtained experimentally, which describe the properties of a continuous medium. In a microscopic approach the macroscopic conservation laws are derived from the Boltzmann equation, according to the Chapman-Enskog procedure. The transport coefficients are then obtained in terms of molecular parameters. When only binary collisions are taken into account the following expressions are valid in a mixture of dilute gases[11]

$$D = \frac{3}{8 N \sigma_{12}^2} \left(\frac{T k_B (m_1 + m_2)}{2 \, m_1 m_2} \right)^{1/2} = 2.628 \times 10^{-3} \frac{\left(T^3 (M_1 + M_2) / 2 M_1 M_2 \right)^{1/2}}{P \sigma_{12}^2} \quad \text{(rigid sphere)}$$

$$= 2.628 \times 10^{-3} \frac{\left(T^3 (M_1 + M_2) / 2 M_1 M_2 \right)^{1/2}}{P \sigma_{LJ12}^2 \, \Omega_{12}^{11} (T_{12}^*)} \, , \qquad \text{(Lennard-Jones pot.)} \quad (17)$$

$$\eta = \frac{\eta_1 \eta_2}{\left(c' \, \eta_2^{1/2} + (1-c') \, \eta_1^{1/2} \right)^2} \qquad \qquad (18)$$

$$\eta_1 = \frac{1}{4 \sigma_1^2} \left(\frac{k_B m_1 T}{\pi} \right)^{1/2} = 2.6693 \times 10^{-5} \frac{(M_1 T)^{1/2}}{\sigma_1^2} \, , \quad \text{(rigid sphere)}$$

$$= 2.6693 \times 10^{-5} \frac{(M_1 T)^{1/2}}{\sigma_{LJ1}^2 \, \Omega^{(22)} (T^*)} \, , \qquad \text{(Lennard-Jones pot.)}$$

$$K'_P = \frac{c' (1-c') \, N \, (m_2 - m_1)}{\rho} \, . \qquad \qquad (19)$$

Here σ_i and σ_{ij} are molecular diameters in the rigid sphere model, σ_{LJi} and σ_{LJij} are parameters describing the Lennard-Jones interaction potential and $\Omega^{(ij)}$ (T_{12}^*) are Chapman-Cowling integrals as a function of the reduced temperature.

For very dilute gases the equations lose their validity, because the variation of the dynamical variables over a mean free path becomes large. The dimensionless constant which is characteristic for the transition from the hydrodynamic to the kinetic regime may be written in the form,[12]

$$y = \sqrt{8\pi} \ \frac{N\sigma^2}{k} \ = \ \frac{\lambda}{\pi^{3/2}\bar{\ell}} \ , \tag{20}$$

where λ is the wave length of periodic variations and $\bar{\ell} = 1/\sqrt{2}\pi N\sigma^2$ is the mean free path. For $y > 3$ the hydrodynamic equations are valid. To be sure that we are in the hydrodynamic regime, we have calculated the parameter y for various gases in table I.

Table I. Gas Parameters for a Rigid Sphere

Model at $T = 300°K$

	σ (Å)	M	$\eta \times 10^5$ g/sec-cm	$\bar{\ell}$	y
H_2	2.731	2.016	8.986	.1123/P	.5548P
H_e	2.174	4.003	19.821	.1770/P	.3520P
X_e	4.916	131.3	23.372	.0346/P	1.800P
SF_6	6.323*	146.05	13.974	.0209/P	2.978P

*Calculated from the measured value of η

σ is hard sphere collision diameter of the molecule in angstrom

M is molecular weight

$\bar{\ell}$ is mean free path in microns

η is the shear viscosity

P is the pressure in atmospheres

III. Coupling with the Electromagnetic Field

In the presence of an electric field the differential free energy per unit mass has to be augmented by terms due to variations in the electric field and the dielectric constant and takes on the form,

$$dF = - SdT + \mu_o dc + P \rho^{-2} d\rho - \frac{\epsilon \underline{E} \cdot d\underline{E}}{4 \pi \rho} - \frac{E^2 d\epsilon}{8 \pi \rho} \quad . \tag{21}$$

Here ϵ must be regarded as a function of the variables T, c and ρ.

It can be shown that the force per unit volume for an uncharged dielectric is changed in the presence of an electric field by an amount,[13]

$$\rho \nabla \left(\frac{1}{8 \pi} E^2 \left(\frac{\partial \epsilon}{\partial \rho} \right)_{T,c} \right) - \frac{E^2}{8 \pi} \left(\frac{\partial \epsilon}{\partial c} \right)_{\rho,T} \nabla c - \frac{E^2}{8 \pi} \left(\frac{\partial \epsilon}{\partial T} \right)_{c,\rho} \nabla T \quad .$$

The last two terms are usually negligible compared to the first term. The effect of electrostriction is therefore to add a term $\frac{1}{8 \pi} \nabla \left(E^2 \frac{\partial \epsilon}{\partial \rho} \right)$ to the right hand side of Eq. (14). For an isotropic fluid without macroscopic flow, so that the nonlinear hydrodynamic term $(\underline{V} \cdot \nabla) \underline{V}$ may be ignored, the combination of Eqs. (13) and (14) leads to a small amplitude sound wave equation in a linearized form,

$$\frac{\partial^2 \rho}{\partial t^2} = \nabla^2 P - \frac{\rho_o}{8 \pi} \nabla^2 \left(\left(\frac{\partial \epsilon}{\partial \rho} \right)_{T,c} E^2 \right) + \frac{1}{\rho_o} \left(\frac{4}{3} \eta + \zeta \right) \nabla^2 \left(\frac{\partial \rho}{\partial t} \right) \quad . \tag{22}$$

In this equation ρ may be expressed in terms of the independent variables P, T and c by means of the equation of state

From Eq. (21) it is clear that the entropy per unit mass in the presence of an electric field is changed by an amount $\frac{E^2}{8 \pi \rho} \left(\frac{\partial \epsilon}{\partial T} \right)_{P,c}$

In a non-absorbing dielectric the thermal diffusion equation requires a correction term $\dfrac{T}{8\pi\rho\,c_p}\left(\dfrac{\partial\epsilon}{\partial T}\right)_{P,c}\dfrac{\partial E^2}{\partial t}$. In the presence of an optical absorption coefficient α_{opt}, one has to add another term, so that the thermal diffusion equation becomes,

$$\frac{\partial T}{\partial t} + \frac{T}{c_p}\left(\frac{\partial S}{\partial P}\right)_{T,c}\frac{\partial P}{\partial t} - \frac{K_T}{c_p}\left(\frac{\partial\mu}{\partial c}\right)_{P,T}\frac{\partial c}{\partial t} = \chi\,\nabla^2 T - \frac{T}{8\pi\rho\,c_p}\left(\frac{\partial\epsilon}{\partial T}\right)_{P,c}\frac{\partial E^2}{\partial t} \tag{23}$$
$$+ \alpha_{opt}\frac{c_o n E^2}{8\pi\rho\,c_p}$$

In a similar manner the concentration diffusion equation has to be augmented, because the chemical potential per unit mass is changed by an amount $\dfrac{1}{8\pi\rho}\left(\dfrac{\partial\epsilon}{\partial c}\right)_{P,T}E^2$. Instead of Eq. (16), the concentration diffusion equation becomes,

$$\frac{\partial c}{\partial t} = D\left[\nabla^2 c + \frac{K_T}{T}\nabla^2 T + \frac{K_P}{P}\nabla^2 P\right.$$
$$\left. -\{8\pi\rho\left(\frac{\partial\mu}{\partial c}\right)_{T,P}\}^{-1}\nabla^2\{\left(\frac{\partial\epsilon}{\partial c}\right)_{P,T}E^2\}\right] \tag{24}$$

To these three equations the wave equation for the electromagnetic field in the isotropic medium must be added,

$$\nabla^2 E - \frac{\epsilon}{c_o^2}\frac{\partial^2 E}{\partial t^2} = \frac{1}{c_\bullet^2}\frac{\partial^2}{\partial t^2}E\left[\left(\frac{\partial\epsilon}{\partial P}\right)_{T,c}P + \left(\frac{\partial\epsilon}{\partial T}\right)_{P,c}T + \left(\frac{\partial\epsilon}{\partial c}\right)_{T,P}c\right] \tag{25}$$

Solutions to the set of four simultaneous second order differential equations (22-25) may be found in the parametric linearized approximation, in which the laser field

$$E_L(x, t) = \frac{1}{2}E_L\,e^{ik_L z - i\omega_L t} + cc$$

is considered as a constant parameter. The scattered light is represented
by a backward wave with amplitude E_s, frequency ω_s, and wave number k_s,

$$E_s \ (r,t) = \frac{1}{2} \ E_s \ e^{-ik_s z - i\omega_s t} + cc$$

Assume for the pressure, concentration and temperature variations solutions
of the form,

$$P \ (r,t) = \frac{1}{2} \ P_1 \ e^{+ ikz - i\omega t} + cc$$

$$c \ (r,t) = \frac{1}{2} \ c_1 \ e^{+ ikz - i\omega t} + cc \qquad (26)$$

$$T \ (r,t) = \frac{1}{2} \ T_1 \ e^{+ ikz - i\omega t} + cc$$

Substitute these expressions into Eqs. (22-25) and ignore all terms higher
than linear in the four small variables E_s, p_1, c_1, and T_1. With the phase
and frequency matching conditions (momentum and energy conservation between
the waves), $k = k_L + k_s$, $\omega = \omega_L - \omega_s$, a secular determinant is obtained which
gives a general dispersion relation. Unfortunately this relation is too com-
plicated to afford physical insight and it will not be written out explicitly.
For the special case $E_L = E_s = 0$, absence of the electromagnetic field, the
relation must of course reduce to the description of sound waves in a fluid
mixture.[10] Numerous papers have been devoted to the description of stimulated
thermal and Brillouin scattering of light in a single component fluid where
$c_1 = 0, \partial \epsilon / \partial c = 0$.[14,15,16]

The interest in this paper is directed toward the concentration
scattering in a nonabsorbing gaseous mixture. A solution for this case will
be presented in the next section, after some explicit expressions for the coup-
ling coefficients $\frac{\partial \epsilon}{\partial P}$ and $\frac{\partial \epsilon}{\partial c}$ have been given.

For a substance obeying the Clausius-Mosotti relation,

$$\frac{\varepsilon - 1}{\varepsilon + 2} = \frac{4\pi}{3} \, N \left[c'\alpha_1 + (1 - c')\alpha_2 \right] \ .$$

where α_1 and α_2 are the polarizabilities of the two molecular species, one readily obtains with the aid of Eqs. (2-4),

$$P \left(\frac{\partial \varepsilon}{\partial P} \right)_{c,T} = \rho \left(\frac{\partial \varepsilon}{\partial \rho} \right)_{c,T} = \rho \left(\frac{\partial \varepsilon}{\partial \rho} \right)_{c',T} = \frac{1}{3} \, (\varepsilon - 1) \, (\varepsilon + 2) \ , \tag{27}$$

and

$$\left(\frac{\partial \varepsilon}{\partial c} \right)_{P,T} = \left(\frac{\partial \varepsilon}{\partial c'} \right)_{N,T} \frac{\partial c'}{\partial c} = \frac{4\pi}{9} \, \frac{(\varepsilon + 2)^2 \, m_1 \, m_2 \, N}{[m_2 \, c + (1-c) m_1]^2} \, (\alpha_1 - \alpha_2) \ . \tag{28}$$

For a gas with a small optical density one may put $(\varepsilon + 2) \, / 3 \sim 1$.

It should be noted that the coupling to the concentration can only become comparable to the density coupling for $\alpha_1 \gg \alpha_2$.

IV. Stimulated Concentration and Brillouin Scattering

In a nonabsorbing gaseous mixture the coupling of the temperature with the electric field and the concentration may be ignored, $\alpha_{opt}=0$, $\partial\varepsilon/\partial T=0$ and $K_T=0$, in Eqs. (23-25). The temperature Eq. (23) may be combined with the sound wave equation. For small thermal conductivity χ, this leads to a sound wave propagating with the adiabatic velocity $v_s = \gamma v_T$ instead of the isothermal velocity v_T, and an additional thermal damping term[10]. Here $\gamma = C_p/C_v$ is the ratio of the specific heats at constant pressure and volume respectively. The sound wave equation may thus be written in the form,

$$\frac{\partial^2 P}{\partial t^2} + v_s^2\left(\frac{\partial\rho}{\partial c}\right)_{P,T}\frac{\partial^2 c}{\partial t^2} = v_s^2\,\nabla^2 P + \left[\frac{1}{\rho_o}\left(\frac{4}{3}\eta + \zeta\right) + \chi(\gamma-1)\right]\frac{\partial}{\partial t}(\nabla^2 P)$$
$$-(8\pi)^{-1}\rho_o\left(\frac{\partial\varepsilon}{\partial\rho}\right)_{T,c}v_s^2\,\nabla^2 E^2 - \frac{\rho_o}{4\pi(\varepsilon+2)}E^2\,(\partial\varepsilon/\partial\rho)^2\,\nabla^2 P\ . \tag{29}$$

The last term arises from the gradient of $\partial\varepsilon/\partial\rho$ in Eq. (22), as evaluated by means of Eq. (27). This term was inadvertently omitted by Wang.[17] It leads to an additional contribution of the intensity dependent Brillouin shift.[17]

The Stokes wave equation (25) is reduced to the form,

$$\nabla^2 E_s - \frac{\varepsilon(\omega_s)}{c_o^2}\frac{\partial^2 E_s}{\partial t^2} = \frac{1}{c_o^2}\left(\frac{\partial\varepsilon}{\partial P}\right)_{c,T}\frac{\partial^2(P\,E_L)}{\partial t^2} + \frac{1}{c_o^2}\left(\frac{\partial\varepsilon}{\partial c}\right)_{P,T}\frac{\partial^2(c\,E_L)}{\partial t^2} \tag{30}$$

The concentration equation (24) takes the form,

$$\frac{\partial c}{\partial t} = D\left[\nabla^2 c_1 + \frac{K_P}{P_o}\nabla^2 P\right] - \left\{8\pi\rho_o\left(\frac{\partial\mu}{\partial c}\right)_{T,P}\right\}^{-1}D\left(\frac{\partial\varepsilon}{\partial c}\right)_{P,T}\nabla^2 E^2 \tag{31}$$
$$- \left\{8\pi\rho_o\left(\frac{\partial\mu}{\partial c}\right)_{T,P}\right\}^{-1}D\,E^2\,\nabla^2\left(\frac{\partial\varepsilon}{\partial c}\right)_{P,T}\ .$$

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The last term may be evaluated by means of Eq. (28) in terms of $\nabla^2 p_1$ and $\nabla^2 c_1$. It corresponds to an intensity dependent frequency shift of the stimulated concentration scattering, analogous to a similar term in Eq. (29) for the Brillouin scattering.

The set of three coupled equations (29-31) for E_s, p_1 and c_1 may now be solved by the same procedure as followed by Herman and Gray[15] for the case of thermal Brillouin scattering in a one component fluid, where the three variables were E_s, p_1 or ρ_1 and T_1.

Since the characteristic relaxation times for diffusion and acoustic damping are assumed to be short compared to the laser pulse duration we may look for a steady state solution for E_s. The imaginary part of the wave vector of the Stokes wave due to the driving terms on the right side of Eq. (30) determines the amplitude spatial gain constant of the stokes wave. We thus solve the two inhomogeneous Equations 29 and 31 for p_1^* and c_1^* in terms of E_s and substitute into Eq. (30), using the "ansatz" (26). One thus finds the power gain constant for combined stimulated Brillouin - concentration scattering,

$$G_{B,c} = \text{Im} - \frac{|E_L|^2 \omega_s}{16 \pi c_o n} \left[(Dk^2 + i\omega) \frac{\gamma_e^2 v_s^2 k^2}{P_o} + \frac{\gamma_e v_s^2 \omega^2 \left(\frac{\partial \rho}{\partial c}\right)_{P,T} \left(\frac{\partial \varepsilon}{\partial c}\right)_{P,T} Dk^2}{P_o \rho_o \left(\frac{\partial \mu}{\partial c}\right)_{P,T}} \right.$$

$$+ \frac{\left(\omega_B^2 - \omega^2 + i\omega \eta' \frac{k^2}{\rho_o}\right) Dk^2 \left(\frac{\partial \varepsilon}{\partial c}\right)_{P,T}^2}{\rho_o \left(\frac{\partial \mu}{\partial c}\right)_{P,T}} - \frac{Dk^4 \kappa_P \gamma_e v_s^2 \left(\frac{\partial \varepsilon}{\partial c}\right)_{P,T}}{P_o} \right] \tag{32}$$

$$\times \left[(\omega_B^2 - \omega^2 + i\omega \eta' \frac{k^2}{\rho_o}) (Dk^2 + i\omega) + \frac{v_s^2 \omega^2 Dk^2 \kappa_P \left(\frac{\partial \rho}{\partial c}\right)_{P,T}}{P_o} \right]^{-1} .$$

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The following abbreviations have been used,

$$\omega_B^2 = \left(v_s^2 - \frac{\gamma_e^2 |E_L|^2}{4\pi \rho_o (\varepsilon + 2)} \right) k^2 \tag{33}$$

$$\gamma_e = \rho_o \left(\frac{\partial \varepsilon}{\partial \rho} \right)_{c,T} = \frac{1}{3} (\varepsilon - 1)(\varepsilon + 2)$$

$$\eta' = \frac{4}{3} \eta + \zeta + \rho_o \chi (\gamma - 1)$$

$$\omega = \omega_L - \omega_s \quad , \quad k_L = k + k_s \quad , \quad \omega_s = \frac{c_o k_s}{n} \quad .$$

The expression (32) shows large resonant values of the gain in the vicinity of $\omega = \omega_B$ (stimulated Brillouin gain) and in the vicinity of $\omega \simeq o$ (stimulated concentration gain). The latter may be nearly uncoupled from the Brillouin gain and is approximately given by,

$$G_c = \frac{\omega_s k^2 D (\partial \varepsilon/\partial c)_{P,T}^2 |E_L|^2}{16\pi nc_o \rho_o (\partial \mu/\partial c)_{P,T}} \quad \frac{\omega}{\omega^2 + D^2 k^4} \quad . \tag{34}$$

It assumes a maximum value for a small stokes shift $\omega = D k^2$. We may write $G_{c \; max} = g_c I_L$, where $I_L = c_o n |E_L|^2 / 8\pi$ is the laser intensity and the gain coefficient for stimulated concentration scattering is given by,

$$g_c = \frac{k_s (\partial \varepsilon/\partial c)_{P,T}^2}{4c_o n^3 \rho_o (\partial \mu/\partial c)_{P,T}} \quad . \tag{35}$$

The resonance at $\omega = \omega_B$ leads the usual stimulated Brillouin gain with the maximum gain factor given by,

$$G_{B, \; max} = \frac{\omega_s |E_L|^2 \; \rho_o \left(\frac{\partial \varepsilon}{\partial \rho} \right)^2 \omega_B}{32\pi c_o n \quad v_s^2 \; \Gamma} = g_B I_L \quad , \tag{36}$$

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where

$$g_B = \frac{k_s \; \rho_o\left(\frac{\partial \epsilon}{\partial \rho}\right)^2}{2 c_o n^3 k v_s \eta_T} \tag{37}$$

and

$$\tag{38}$$

$$\Gamma = \frac{k^2 \eta_T}{2\rho_o} = \frac{k^2}{2\rho_o} \left[\frac{4}{3}\eta + \zeta + \kappa\left(\frac{1}{C_V} - \frac{1}{C_P}\right) + \frac{D v_s^2}{\rho\left(\frac{\partial\mu}{\partial c}\right)_{P,T}} \left(\frac{\partial\rho}{\partial c}\right)^2_{P,T} \right].$$

In a binary mixture the sound wave has an extra damping term (last term of Eq. 38) due to the coupling with the concentration fluctuation.[18] This extra damping depends on $\left(\frac{\partial\rho}{\partial c}\right)^2_{P,T}$, and consequently the sound wave is heavily

damped in a mixture with a large difference of component masses. The threshold of stimulated Brillouin scattering is increased by mixing. The ratio of the gain constants for stimulated concentration and Brillouin scattering is,

$$\frac{g_c}{g_B} = \frac{k v_s \eta_T (\partial\epsilon/\partial c)^2_{P,T}}{2 \rho_o^3 (\partial\epsilon/\partial\rho)^2_{c,T}(\partial\mu/\partial c)_{P,T}} \tag{39}$$

From Eqs. (38) and (28) it follows that a large difference in polarizability and a large difference in mass of the two components is favorable to make this ratio appreciable. Furthermore, the total density or pressure should be kept low, because the ratio is inversely proportional to the density. This is a consequence of the fact that g_c is proportional to the density, but g_B increases proportional to the square of the density, as the acoustic damping constant is inversely proportional to the density.[19]

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For a gaseous mixture Eq. (39) can be simplified with the aid of Eqs. (11), (27), and (28) to,

$$\frac{g_c}{g_B} = \frac{k \, v_s \, \eta_T}{2N \, k_B \, T} \qquad \frac{c' \, (1-c') \, (\alpha_1 - \alpha_2)^2}{[\alpha_1 \, c' + (1-c') \, \alpha_2]^2} \qquad (40)$$

with η_T given by Eq. (38).

V. Numerical Results and Comparison with Experiment

If one substitutes typical values for a liquid mixture into Eq. (39), the ratio is found to be very small compared to unity. For example, a mixture of n-hexane (58 percent weight concentration) and nitrobenzene (42 percent) gives $g_c/g_B \sim 1.25 \times 10^{-5}$. This throws doubt on the interpretation of the experiment described in reference 7. Near the critical point in a binary mixture, where $\partial\mu/\partial c \to 0$, the concentration gain could become large. The diffusion constant D approaches zero at the same time. The correlation length and correlation time for the concentration fluctuations becomes very long, $\sim 10^{-3}$ sec. The steady state analysis breaks down for solid state laser pulses, and even for gas laser beams it would be difficult to keep the required coherence for such long times. A transient analysis must be made and the experimental difficulties would be further enhanced by the large spontaneous critical opalescence, although amplification of this spontaneous emission at high intensities may well be detectable.

Even for gaseous mixtures it is difficult to obtain a ratio g_c/g_B comparable to unity. It should be kept in mind that the total pressure cannot be decreased arbitrarily, because the parameter y in Eq. (20) would become too small and the kinetic regime would take over. Also the required path length to obtain appreciable overall gain would be too long.

In table II, we have calculated numerical values of the constants which appear in g_c and g_B as a function of helium concentration, for a gaseous mixture of SF_6 and He at ten atmospheres total pressure and T = 300°K. The diffusion constant D = .03747 cm^2/sec and viscosity are calculated from Eqs. (17) and (18), using the tabulated Lennard-Jones potential parameters (ref.11). The adiabatic sound velocity is calculated using the expression

Table II. Comparison of the Concentration and Brillouin Gain Coefficients

in a Mixture of SF_6 and He at Total Pressure 10 atm.

He c'	$\eta_{mix} \times 10^4$ g/cm sec	$v_s \times 10^{-4}$ cm/sec	$\Delta \nu_B$ (cm^{-1})	$\eta_T \times 10^4$ g/cm sec	g_c/g_B	$g_B \times 10^{10}$ cm/W	$g_c \times 10^{10}$ cm/W
0	1.397	1.509	.0145	2.474	0	53.352	0
.1	1.444	1.608	.0154	5.821	.0076	17.405	.1325
.2	1.492	1.723	.0165	9.305	.0295	8.137	.2396
.3	1.542	1.859	.0179	12.923	.0734	4.220	.3096
.4	1.596	2.025	.0194	16.646	.1568	2.255	.3535
.5	1.652	2.233	.0214	20.449	.30	1.191	.3686
.6	1.711	2.507	.0241	24.392	.596	.5935	.3534
.7	1.773	2.895	.0278	27.932	1.142	.2711	.3097
.8	1.839	3.509	.0337	31.035	2.310	.1037	.2396
.9	1.908	4.731	.0454	31.898	5.067	.0262	.1327
.95	1.945	6.088	.0584	28.495	7.030	.0100	.0703
.98	1.967	7.635	.0733	20.461	5.444	.0053	.0287
1.	1.982	10.194	.0979	4.626	0	.0086	0

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$v_s = [(c'\gamma_1 + (1- c') \gamma_2) k_B T/(c'm_1 + (1 - c') m_2)]^{1/2}$. The high frequency specific heat ratio for He and SF_6 are taken to be $\gamma_1 = 1.667$ and $\gamma_2 = 1.333$.[20] η_T and the gain coefficient and ratio g_B, g_c/g_B are calculated from Eqs. (37), (38) and (40) respectively.

We have also calculated the gain coefficients g_B and g_c for a mixture of five atmospheres of SF_6 as a function of the additional helium pressure. The results are plotted in Figure 1. It is seen that stimulated concentration scattering may be expected to dominate when the concentration of helium is around 0.85. The gain ratio becomes larger mainly because the damping of sound wave increases and the Brillouin gain becomes smaller. Thus, in order to be able to see the stimulated concentration scattering one has to increase the laser power from the value used in obtaining the stimulated Brillouin scattering. The expected frequency behavior of the stimulated back-scattered light has been plotted in Figure 2. The frequency shift should increase for the stimulated Brillouin scattering when helium is added in SF_6. As the concentration gain becomes larger than the Brillouin gain one would see a sudden decrease in the frequency shift.

The numerical results for mixtures of helium and Xenon gas are given in table III. These results should be compared with the data reported in reference 8. The experiment in reference 8 was done for a Xe and He mixture with He concentration $c' = 0.9$. At this concentration the Brillouin shift should be 0.0479 cm^{-1} instead of 0.055 cm^{-1} reported. According to table III the gain ratio at a total pressure of 3 atm should be $g_c/g_B = 21$ in contrast to $g_c/g_B = 0.1$ as estimated in reference 8. The observed concentration shift 0.033 ~ 0.042 cm^{-1} agrees with the value $\Delta v_c = \dfrac{Dk^2}{2\pi c} = 0.032$ cm^{-1}.
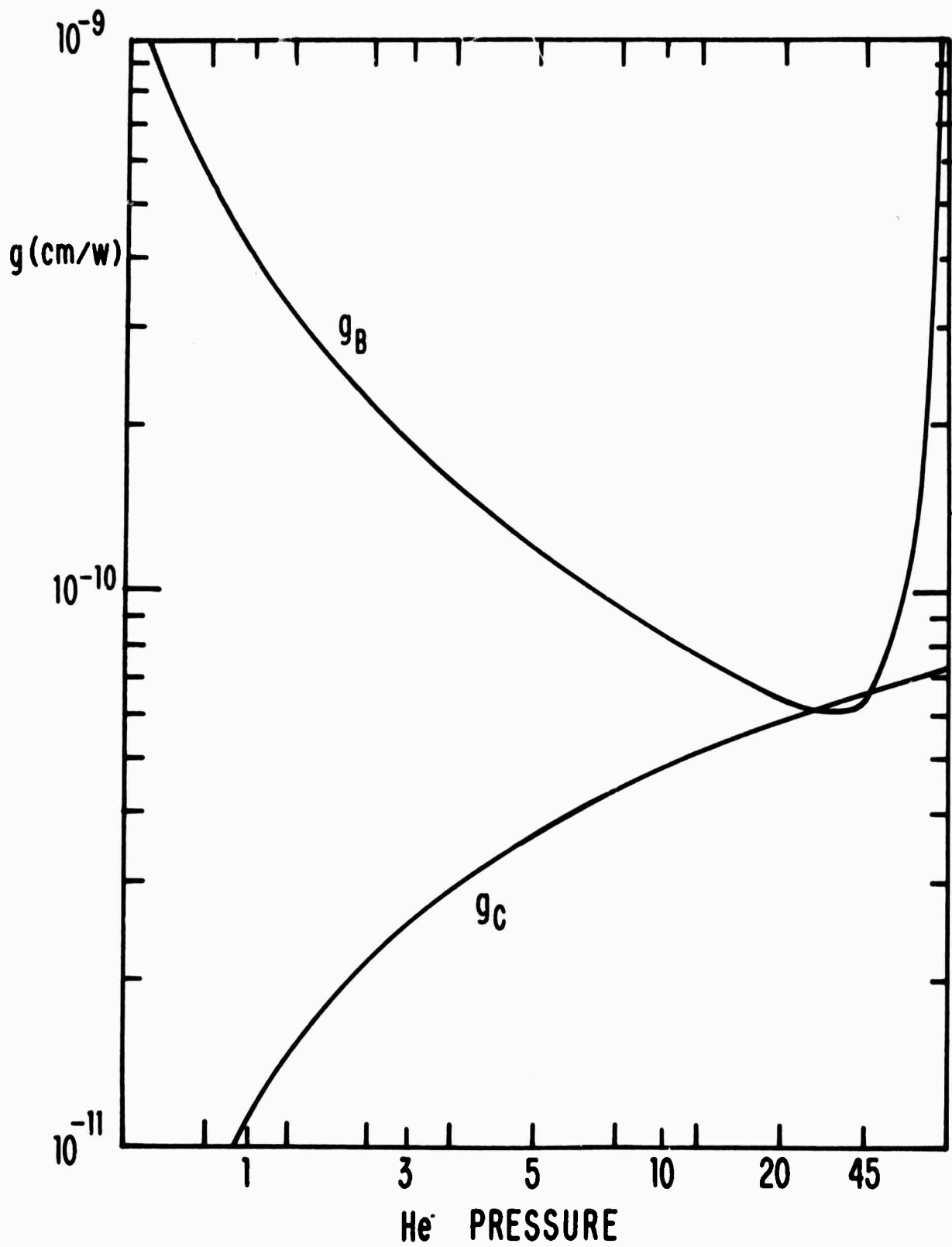
Fig. 1. The backward Brillouin and concentration gain coefficients as functions of He partial pressure (in atm.) keeping the partial pressure of SF_6 at 5 atm.
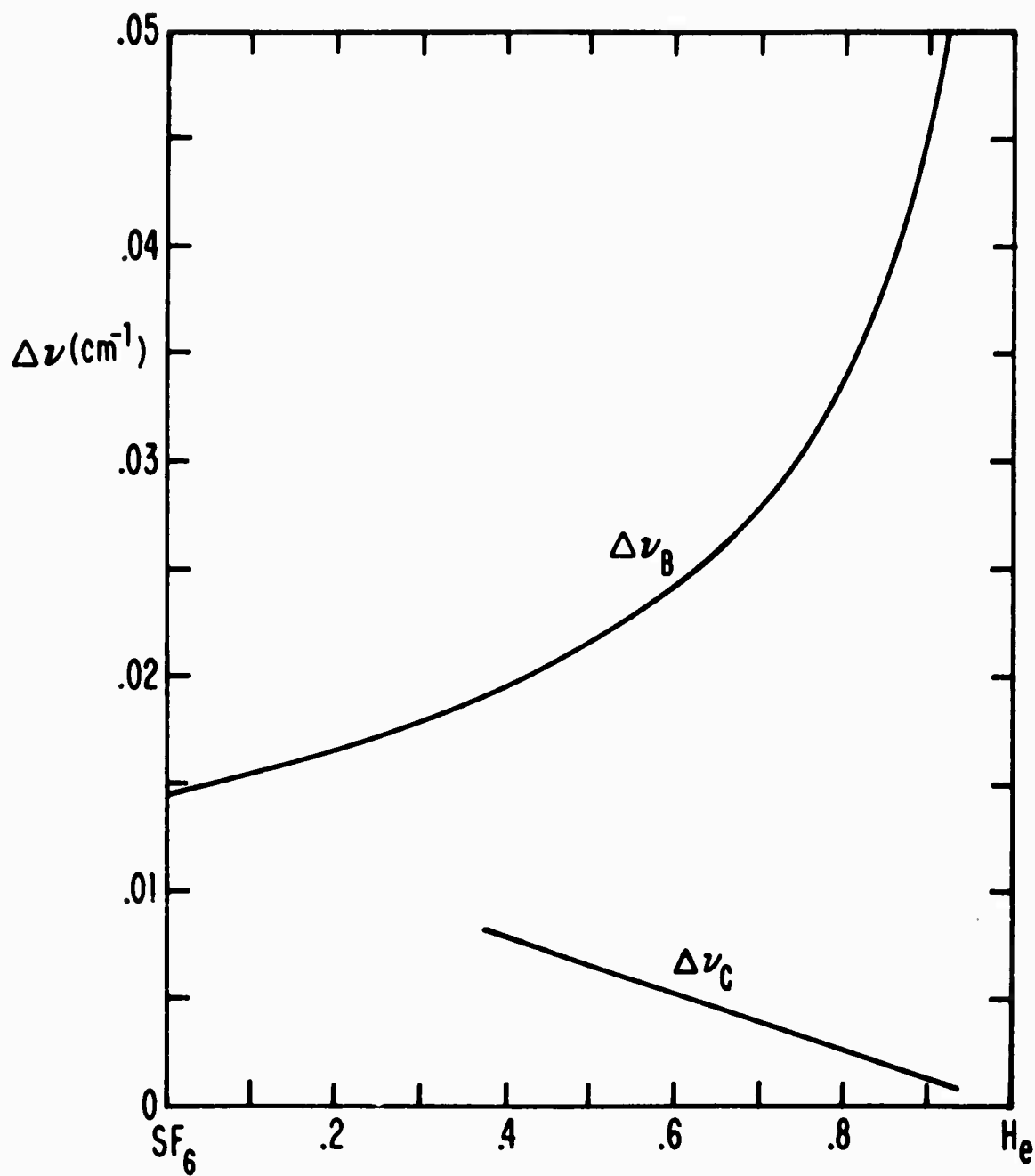
Fig. 2.   The Stokes frequency shift of the backward scattered light from
a mixture of SF_6 and He as a function of He concentration,
starting with five atmosphere SF_6.

Table III. Comparison of the Concentration and Brillouin Gain Coefficients

in a Mixture of Xe and He at Total Pressure 10 atm.

He c'	$\eta_{mix} \times 10^4$ g/cm sec	$v_s \times 10^{-4}$ cm/sec	$\Delta \nu_B$ (cm^{-1})	$\eta_T \times 10^4$ g/cm sec	g_c/g_B	$g_B \times 10^{10}$ cm/W	$g_c \times 10^{10}$ cm/W
0	2.337	1.779	.0171	5.453	0	16.830	0
.1	2.296	1.872	.0180	10.372	.0157	6.890	.1078
.2	2.257	1.981	.0190	15.254	.0541	3.546	.1916
.3	2.219	2.113	.0203	20.082	.1279	1.968	.2516
.4	2.183	2.274	.0218	24.824	.2586	1.112	.2876
.5	2.147	2.478	.0238	29.428	.4853	.6173	.2996
.6	2.112	2.751	.0264	33.800	.8855	.3247	.2875
.7	2.078	3.138	.0301	37.745	1.6262	.1547	.2516
.8	2.044	3.755	.0361	40.677	3.1129	.0615	.1914
.9	2.012	4.984	.0479	40.322	6.3119	.0170	.1073
.95	1.996	6.117	.0587	34.871	7.821	.0073	.0569
.98	1.986	7.792	.0748	24.347	5.731	.0041	.0235
1.	1.982	10.193	.0978	4.624	0	.0086	0

At a higher total pressure the stimulated Brillouin should take over the stimulated concentration scattering as suggested in reference 8, however this should not occur at a total pressure as low as 4 atmospheres. In conclusion, the theoretical calculations presented in this paper are in partial agreement with the observation in reference 8, however the agreement is not conclusive. Further experimental investigation of the stimulated concentration scattering as a function of concentration and total pressure of the mixture is required. Preliminary observations on SF_6-He mixture[9] have shown that the onset of stimulated concentration scattering can occur at a rather high total pressure. It is clear that in problems relating to the propagation of intense light beams in the atmosphere, stimulated concentration scattering may be ignored.

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The parameter y characterizes the transition of the kinetic to the hydrodynamic regime of a single component fluid, the transition between these regimes in a mixture is yet to be investigated. However, one can expect that if one component is in the hydrodynamic regime with its partial pressure the mixture should be in the hydro-dynamic regime.

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$\Delta v_s^2 = - (5 \epsilon + 2) \rho_0 (\partial \epsilon / \partial \rho)^2 |E_\ell|^2 / 16 \pi (\epsilon + 2) \epsilon$. This makes the agreement with the experimental breakdown intensity even better. For gases with high polarizabilities such as SF_6 and Xe, the intensity dependent effect is comparatively small. We shall neglect this effect in the following calculations.

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APPENDIX B

OBSERVATION OF STIMULATED CONCENTRATION SCATTERING[*]

IN A MIXTURE OF SF_6 AND He

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ABSTRACT

→Stimulated Rayleigh scattering caused by concentration fluctuations in a non-absorbing binary gaseous mixture has *is discussed,* been observed. The experimental results are in good agreement with theoretical expectations. ( )

Stimulated Rayleigh scattering due to temperature fluctuations has been observed in liquids and gases, especially when light absorption is present.[1-3] Stimulated Rayleigh wing scattering from anisotropic molecules was observed even earlier.[4,5] The possibility of observing the stimulated Rayleigh scattering from concentration fluctuations in a binary mixture of fluids was first suggested by Bespalov and Kubarev.[6] Their observation of stimulated Rayleigh scattering in a binary liquid mixture probably has a different origin, since a detailed theoretical analysis of stimulated concentration scattering[7] shows that the stimulated concentration gain coefficient in the liquid mixture should be several orders of magnitude smaller than the stimulated Brillouin gain. Aref'ev and Morozov[8] have reported the observation of stimulated concentration scattering in a mixture of three atmospheres of helium and 0.3 atmospheres of xenon. The interpretation of their experimental results is, however, inconclusive for the following reasons: a) The quoted pressures are so low that the hydrodynamic theory of concentration fluctuations is not valid and a kinematic theory should be used. b) The effect was not observable at higher pressures, in disagreement with the theory of stimulated concentration scattering. c) The effect was observable only with the simultaneous presence of a spark discharge in the gas cell. This may cause thermal Brillouin and Rayleigh scattering to occur.[9]

It is the purpose of this note to report experimental results which are in good agreement with the theory of stimulated concentration scattering. A mixture of SF_6 and He was chosen in our experiments, because the theory shows that a large difference in polarizability and mass is required, if the stimulated concentration gain is to become comparable to the Brillouin gain.[7] Furthermore, the totally symmetric molecule SF_6 avoids the competition from Rayleigh wing scattering and selffocusing caused by molecular reorientation. It is important to keep the total pressure low because the Brillouin gain coefficient g_B increases proportional to the square of the pressure[10] and the stimulated concentration gain coefficient g_c increases proportional to the pressure itself, at a given relative concentration. The partial pressures of the two components of the mixtures studied were 5 atm. SF_6 and 0 to 33 atm. He. The stimulated Brillouin effect was observable in the pure SF_6 with pressure of 5 atm. at room temperature.

In our first experiments the decrease in g_B, when He is added to SF_6, was observed as an increase in threshold power to obtain backward scattered Brillouin radiation. This is caused by the drastic increase in damping of hypersonic waves in a mixture with unequal masses. When the partial He pressure exceeded that of SF_6, a decrease in the Stokes shift of the backward scattered light was noted. Since the threshold for stimulated concentration oscillations was very high

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and the concentration stokes shift is small, there were experimental difficulties in controlling the frequency spectrum of the ruby laser and avoiding discharge plasma formation. More definitive results were obtained with the gas amplifier cell arrangement shown in figure 1.

A low power ruby laser oscillator, followed by a ruby laser amplifier yielded 100 MW of power in a 20 nanosecond pulse with a spectral width of .005 cm^{-1}. The collimated beam with cross section of 1 cm^2 is focused into the gas cell by a lens with 25 cm. focal length. In mixtures with He concentration greater than 40%, no stimulated oscillations occurred under these conditions. After passage through the gas cell, the ruby light beam was recollimated and then focussed into a cell containing nitrobenzene. Nitrobenzene has a continuum backward scattered spectrum[6] with a Stokes shift between 0 and 0.6 cm^{-1} due to Rayleigh and Rayleigh-wing scattering with the superposition of a strong stimulated Brillouin signal with a Stokes shift of 0.2 cm^{-1}. This spectrum provides a suitable input signal for the gas cell which can amplify light with Stokes shifts between .019 cm^{-1} and .005 cm^{-1} depending upon the He concentration of the mixture, and does not amplify the nitrobenzene Brillouin component. The power gain in the gas amplifier cell was on the order of e^4. This low gain has the advantage of more nearly satisfying the condition for steady state amplification during the pulse. With characteristic time $\tau = 0.5 \times 10^{-8}$ sec. for the concentration fluctuations, steady state conditions should
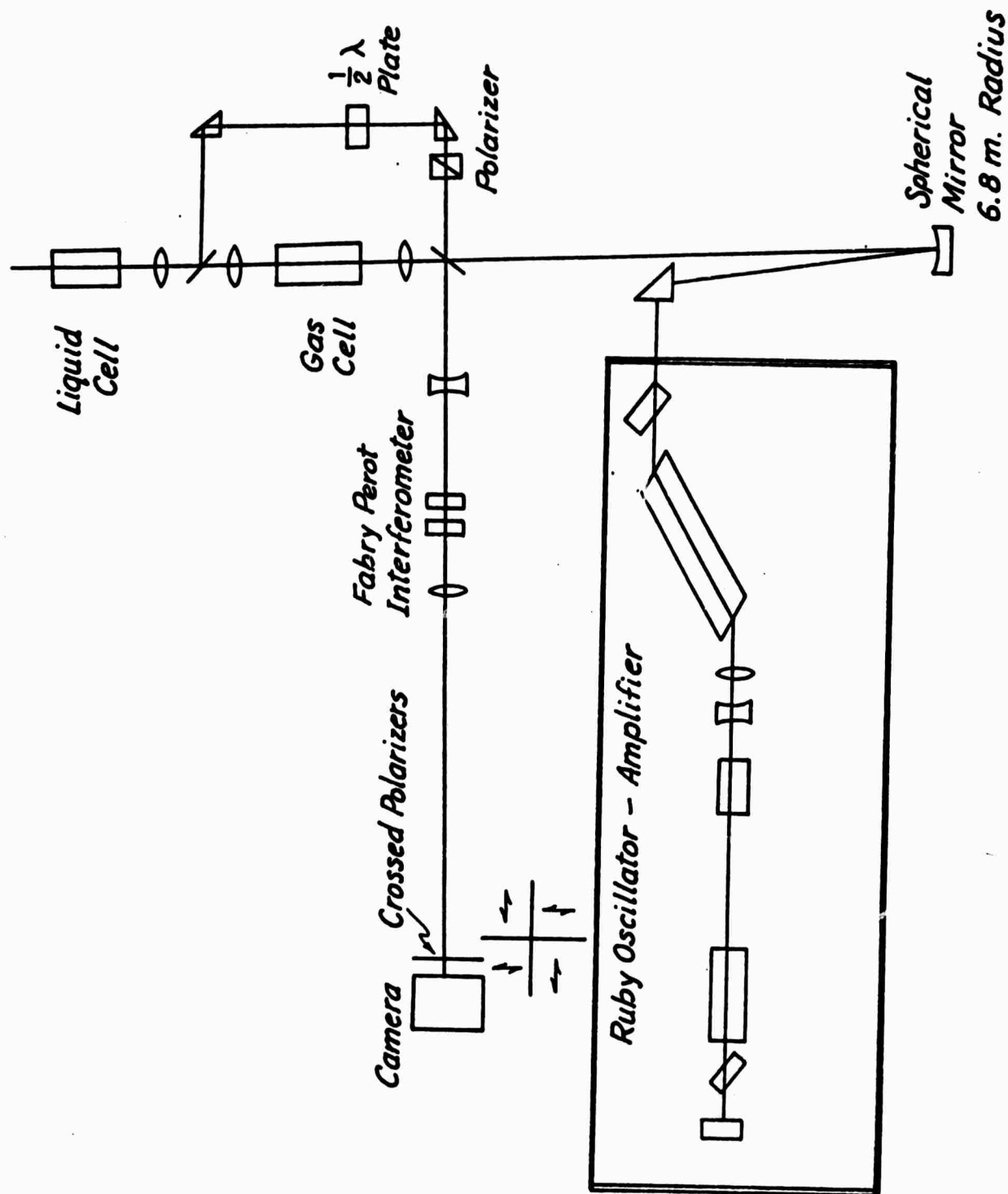
Fig. 1. Schematic of the experimental arrangement to detect amplification by stimulated concentration scattering.

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be approached during a pulse of 2×10^{-8} sec. duration.

The input and output signal of the gas cell were spectrally analyzed in Fabry-Perot interferometers with free spectral ranges of 0.5 cm^{-1} and 0.083 cm^{-1}. The input and output signals were distinguished in the usual way by orthogonal polarizations[1] and an example of the experimental data taken with the low resolution Fabry-Perot is shown in figure 2. Attenuated ruby laser light and nitrobenzene Brillouin light provided calibration rings.

It was observed with the high resolution Fabry-Perot that the frequency of the Stokes shifted light which had the maximum gain in the gas cell changed as a function of the partial helium-pressure in the manner shown in figure 3. At low helium pressures the amplified Stokes shift corresponds to the Brillouin shift in the gaseous mixture and increases somewhat with increasing helium content in good agreement with the theory of the Brillouin effect. The theoretical curves for the adiabatic and isothermal effect are drawn for comparison. We do not wish to draw any conclusions about which of these regimes should apply, but attention should be directed to the relative trend of the Stokes shift of the amplified light. When the partial helium pressure exceeds 8 atmospheres, while the SF_6 partial pressure is kept constant at 5 atmospheres, the Stokes shift of the amplified light decreases rapidly away from the Brillouin shift, and approaches the very small value of .005 cm^{-1} characteristic of the stimulated concentration scattering. The cross-over occurs precisely

— NITROBENZENE BRILLOUIN
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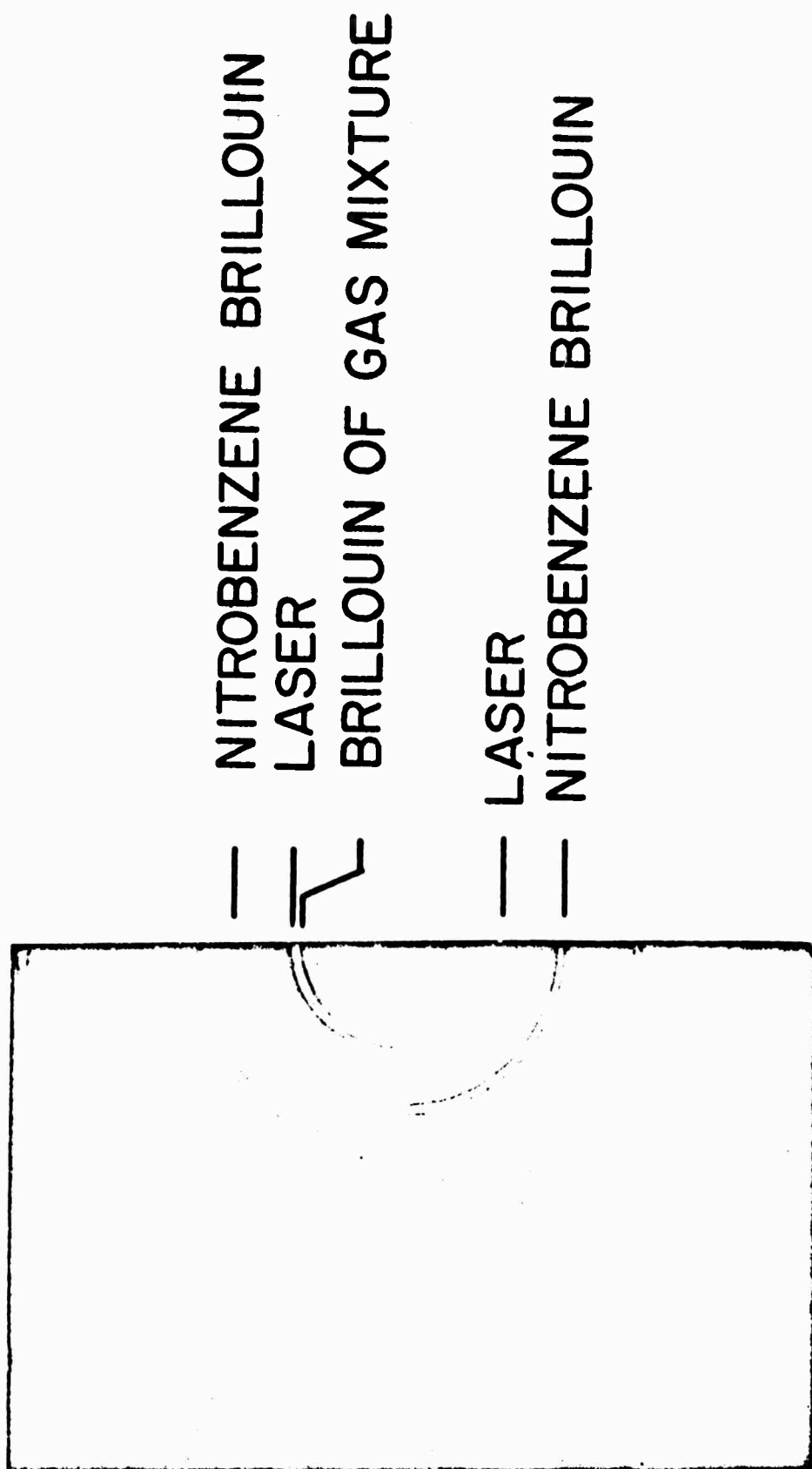— BRILLOUIN OF GAS MIXTURE

— LASER
— NITROBENZENE BRILLOUIN

Fig. 2. Photograph showing the spectral characteristics of the gain in a gas cell, containing partial pressures of 5 atm. SF_6 and 2 atm. He. The lower quadrant represents the spectral quality of the input signal, the upper quadrant represents the spectral quality of the output signal. There is amplification at the Brillouin shift for the gas mixture but not at the laser and nitrobenzene Brillouin reference rings, which provide a frequency calibration. (Unfortunately the reproduction does not show the input intensity near the laser frequency. Experimentally the Brillouin gas line disappears when the nitrobenzene cell is removed.
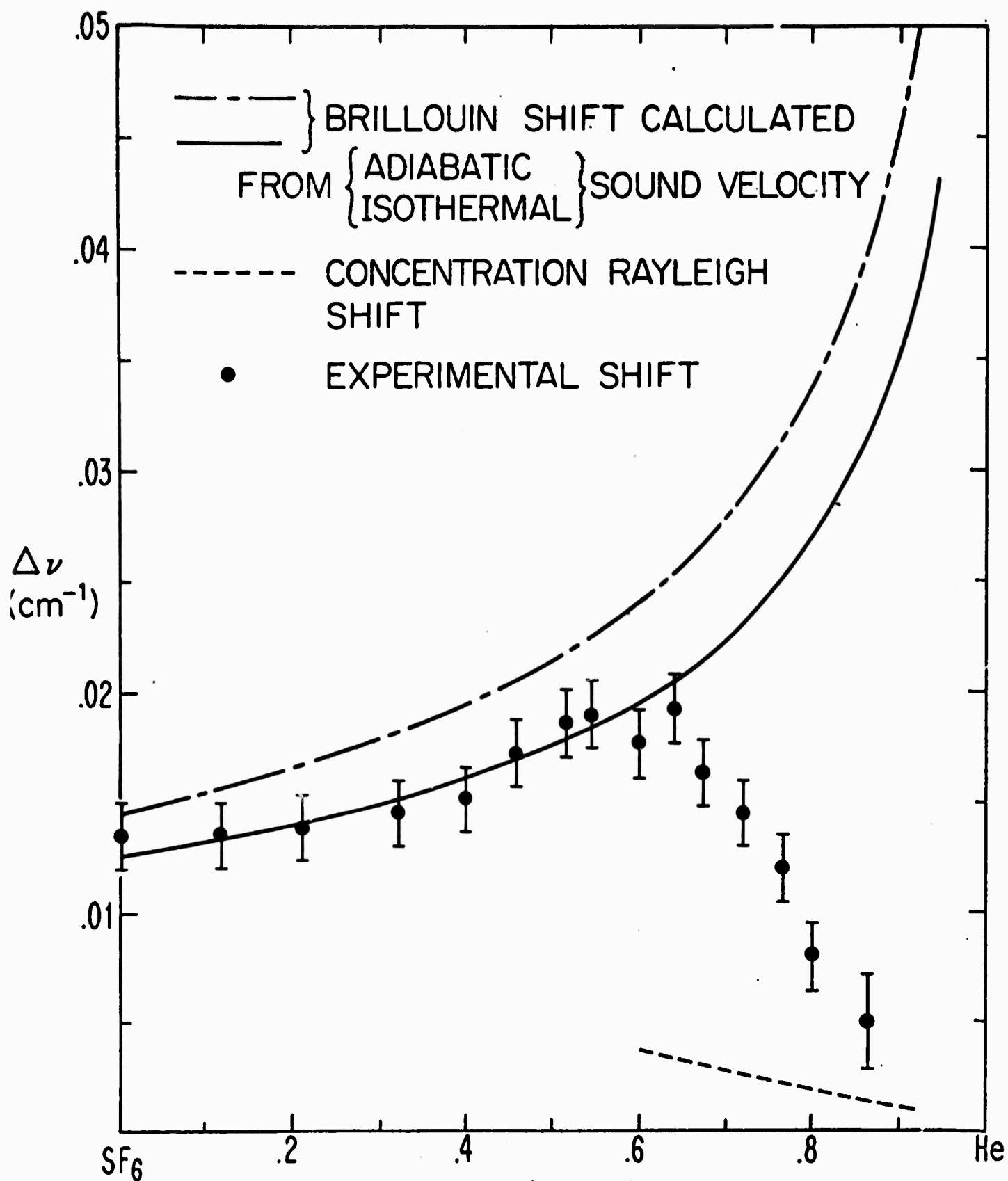
Fig. 3. The Stokes frequency shift of the amplified light in a gas cell with a partial pressure of 5 atm. SF_6, as a function of He concentration. The drawn curves are theoretically calculated Stokes shifts for the pure Brillouin and pure concentration mode.

in the region where the ratio g_c/g_B is about unity according to the theory.[7] It should be noted that the hypersonic and the concentration mode are heavily damped and overdamped respectively at these compositions of the gas mixture. The theory does not permit a clean separation in a pure Brillouin and pure concentration mode as suggested by the drawn lines in figure 3. The experimental results are in good agreement with the existence of a mixed mode transition from dominant stimulated Brillouin scattering to dominant stimulated concentration scattering.

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